Earth Surface Processes and Landforms
Earth Surf. Process. Landforms **24**, 725–730 (1999)

AN INVESTIGATION INTO THE EFFICIENCY OF PARTICLE SIZE SEPARATION USING STOKES' LAW

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Received 7 August 1998; Revised 16 November 1998; Accepted 20 January 1999

ABSTRACT

The accuracy of gravimetric fractionation as a means of obtaining size fractions from marine sediments has been explored. Analysis of the particle size distribution and sediment properties of fractions obtained using this method was undertaken. This has highlighted the extent to which experimental artefacts rather than variations in sediment characteristics may adversely affect the efficiency of the fractionation process. Copyright © 1999 John Wiley & Sons, Ltd.

KEY WORDS: particle size fractionation; laser granulometry

INTRODUCTION

Trace metals and radionuclides are present in greater concentrations in association with fine-grained particles as a result of the greater surface area available for adsorption, this relationship being well documented in both marine and terrestrial environments (Ackermann,1983; Assinder *et al.*,1993). A range of techniques has therefore been developed to physically separate sediment in the silt and clay range ($>4\phi$; $< 63\ \mu$m) on a size-specific basis in order to study the distribution and fate of adsorbed contaminants. These include centrifugation (Ducaroir and Lamy,1995), elutriation (Horowitz and Elrick,1986; Walling and Woodward,1993), heavy-liquid flotation (Cotter-Howells,1993), magnetic separation (Bulman *et al.*,1984) and sieving (Mundschenk,1996). Gravimetric settling is, however, the most commonly employed method; it involves removal of a specific volume of material settling in suspension which, according to Stokes' Law, should contain particles finer than a specific diameter, thereby allowing removal of fractions at predetermined size class intervals. This method allows size fractions to be quickly obtained and involves minimal capital outlay, leading to its widespread use in studies of trace element distribution (Livens and Baxter,1988; Cundy and Croudace,1995; He and Walling,1996).

However, the assumptions implicit within Stokes' Law, that spherical grains of a known uniform density are settling freely in non-turbulent fluid of a constant temperature, will evidently seldom be met even under laboratory conditions, leading to a longstanding debate on the efficiency of the settling procedure (Gibbs,1972). However, it is only recently that advances in laser granulometry as a particle sizing technique have permitted the accurate analysis of size fractions obtained using the settling technique (Walden and Slattery,1993). The present study aims to follow up this research through a detailed examination of size fractions obtained from marine sediments and of the extent to which properties such as sediment density and organic carbon content may influence the observed grain size distribution.

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Contract/grant sponsor: BNFL

CCC 0197-9337/99/080725–06 $17.50

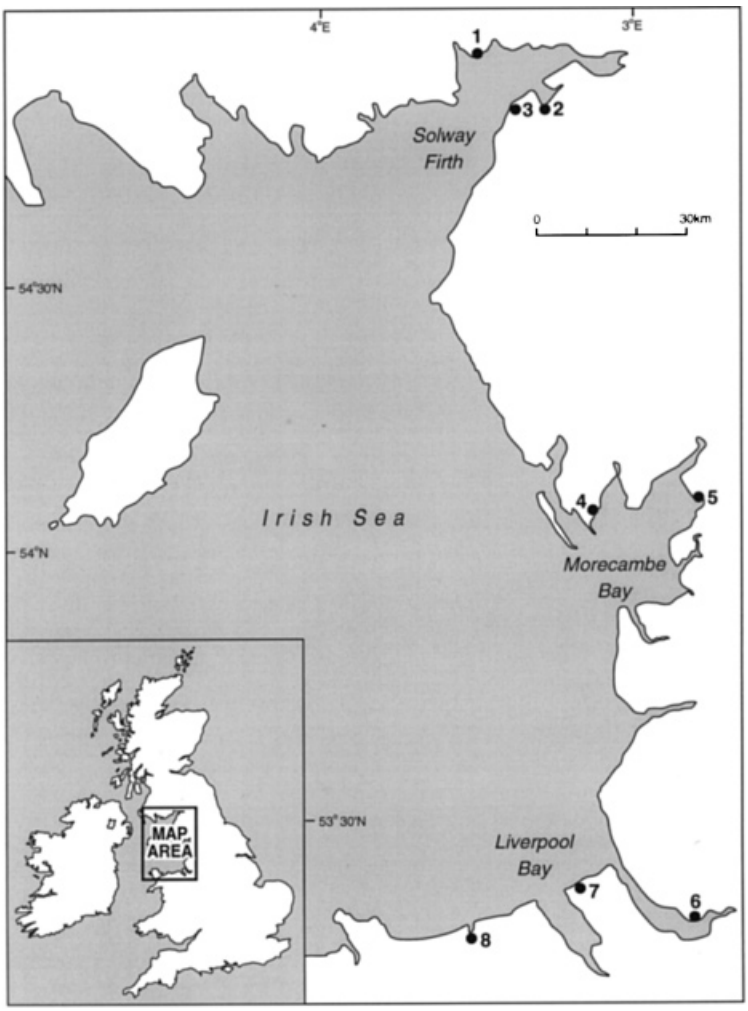

Figure 1. Sampling locations in the eastern Irish Sea

MATERIALS AND METHODS

Eight locations in the eastern Irish Sea, representing a diverse suite of fine-grained salt marsh and intertidal mudflat environments, were visited in March 1995 (Figure 1). Approximately 1 kg of surface sediment was sampled to a depth of 10 mm at each site and frozen within 48 hours of collection.

Representative sub-samples weighing approximately 50 g were taken from each sample and suspended in 300 ml of distilled water. These were dispersed through the addition of 20 ml aliquots of Calgon solution, following which the samples were placed in an ultrasonic bath for 30 min and wet-sieved at 4ϕ.

The density of the sediment finer than 4ϕ obtained from each sample was determined prior to fractionation in order to calculate settling times reflecting variations in sediment composition. The Soil Survey method (Avery and Bascomb,1974) was used for this purpose, in which sediment density is calculated using the difference in volume between a flask filled with distilled water containing a known mass of dry sediment and the volume of the same flask containing distilled water alone. The precision of this technique was verified through duplicate measurements of all samples, which yielded results within 5 per cent of the original data.

The sediment finer than 4ϕ from each sample was again dispersed following the above procedure and decanted into individual glass settling tubes (140 cm $\times$ 15 cm), maintaining a particle concentration of less

than 1 per cent by volume in order to minimize particle collisions during the fractionation process (Galehouse,1970).

Immediately prior to size fractionation, each tube was shaken end over end for 2 min to fully disperse the sediment. Following predetermined settling times, a peristaltic pump was used to remove the topmost 10 cm of suspension; this technique allowed minimal disturbance to the settling suspension. Each tube was then topped up to 1000 ml with distilled water and the operation repeated until the suspension withdrawn became clear, indicating that complete removal of each fraction had been achieved. The temperature of the suspension in all settling tubes ranged from 18 to 21 °C during the experiment.

Size fractions withdrawn using this procedure were analysed directly with no further pretreatment in order to determine the grain size distribution of each fraction. A minimum of four measurements of each size fraction withdrawn was carried out during the settling process in order to detect any temporal changes in grain size distribution. A Malvern Mastersizer S laser granulometer configured to analyse sediment in the $0{\cdot}1$–14ϕ (900–0·05 μm) range in one measurement was used for all particle size analyses. Whilst earlier generations of this instrument have been critically evaluated (Syvitski *et al.*,1991), no comparative assessment of this model has yet been made. However, the difference between the measured and nominal diameter of five standard reference materials ranging from 2·01 μm to 202 μm was less than 5 per cent (Clifton,1998), indicating an acceptable degree of instrumental accuracy. The laser granulometer calculates the percentage of sediment found to lie in successive size classes. Precision of measurement was therefore ensured through undertaking duplicate analyses of each sample until the proportion of sediment recorded in each size class differed by less than 1 per cent.

Size fractions were dried at 40°C and organic carbon content measured following the modified wet oxidation procedure as described by Loring and Rantala 1992. Reference materials were utilized to calibrate individual batches of samples, allowing a standard deviation of 0·045 per cent to be applied to all organic carbon data.

Statistical analyses were undertaken using the non-parametric Spearman rank correlation test in order to identify any relationships between measured sediment properties and size distribution characteristics. This test was selected as the assumptions of normality implicit in the use of parametric statistical tests could not be ensured.

RESULTS AND DISCUSSION

Density

The measured density values ranged between 1·66 and 2·99 g cm^{-3}, which serves to highlight the extent to which the assumption of a constant sediment density may adversely affect the accuracy of gravimetric fractionation. Despite the wide range in organic carbon content measured prior to fractionation (0·95–5·47 per cent), no correlation with sediment density was evident ($r_s = -0{\cdot}1$, $\rho = >0{\cdot}05$). Mineralogical differences reflecting the diverse origin of these samples may, therefore, exert the dominant influence upon density.

Monitoring of particle size distributions

Table I indicates that the mean grain size of fractions obtained from all samples using this procedure was within the required size range. Furthermore, no temporal trend in mean grain size could be identified. This implies that the size distribution of samples obtained *via* fractionation is not dependent upon sediment concentration. Therefore, flocculation was not a significant factor affecting fractionation in this experiment, as particle aggregation at higher sediment concentrations would be manifest in an increase in the mean grain size of sediment removed in the initial stages of fractionation.

Efficiency of size fractionation procedure

Table II summarizes the efficiency of the fractionation procedure using data from particle size analyses of all individual fractions. This reveals that the proportion of sediment lying in the desired size range in each

Table I. Range of mean grain sizes measured in size fractions from all samples

	Size fraction				
(phi)	4–5	5–6	6–7	7–9	>9
(μm)	32–63	16–32	8–16	2–8	<2
Mean grain size (ϕ)	4·0–4·1	5·4–5·7	5·8–6·0	6·4–6·9	8·8–9·1

Table II. Average percentage of sediment by volume coarser, within and finer than required size range measured in size fractions from all samples

	Size fraction				
(phi)	4–5	5–6	6–7	7–9	>9
(μm)	32–63	16–32	8–16	2–8	<2
% coarser	28·4	24·0	34·2	34·2	35·0
% $\pm$ 1σ	12·9	9·2	11·1	17·6	13·1
% within	45·1	31·3	26·3	40·7	65·0
% $\pm$ 1σ	11·7	8·1	3·8	6·9	13·1
% finer	26·5	44·9	39·6	25·1	–
% $\pm$ 1σ	22·6	16·1	13·5	18·3	–

fraction varied from 26 to 65 per cent by volume, indicating that the efficiency of size fractionation using this method is far from ideal. Whilst the magnitude of the standard deviation values indicates some variation between samples, recovery of sediment in the 5–7ϕ range is least efficient, with the percentage of sediment within the required size range being less than that in the coarse or fine tails of the distribution. The fraction finer than 9ϕ is associated with the maximum abundance of sediment within the required range as there is no lower size limit to this fraction.

Figure 2 illustrates a typical example of the grain size distribution of fractions obtained by the gravimetric separation procedure. This demonstrates that all size fractions obtained using this method exhibit distinct modal peaks which are separated by at least one phi unit, indicating that the settling procedure employed does yield qualitatively different size fractions. This also clarifies the nature of the coarse and fine tails of the size fractions. A fine tail composed of sediment finer than 8ϕ is recorded in size fractions theoretically containing no material finer than 7ϕ. Furthermore, despite the preliminary wet-sieving undertaken, material coarser than 4ϕ is evident in most size fractions from all samples, accounting for almost one-third of the 4–5ϕ fraction in the example illustrated.

Despite the fact that sediment density is recognized as a variable property in this study, the efficiency of this procedure does not represent a significant improvement upon previous fractionation experiments using agricultural topsoils and glacial sediments (Walden and Slattery,1993). This implies that common drawbacks may exist in gravimetric settling procedures which merit discussion. The presence of a fine tail extending to 12ϕ reflects the fact that, in common with previous workers (Oldfield and Yu,1994), complete removal of the finer fractions was not achieved in this study, as the suspension being withdrawn during the fractionation procedure did not become clear after a total of ten individual withdrawals. It was therefore impossible to exclude small amounts of finer-grained material in successive size fractions. This has particularly significant implications with regard to interpreting pollutant data from analysis of size fractions obtained using this method.

Discrete particles of organic matter have been considered to give rise to coarse tails in size fractions owing to the lower density of organic matter and, hence, settling velocity (Barbanti and Bothner,1993). However, despite the wide range of organic carbon content in size fractions measured in this study (0·07–8·01 per cent), Table III indicates that the abundance of coarse tails in size fractions is not related to organic carbon content.

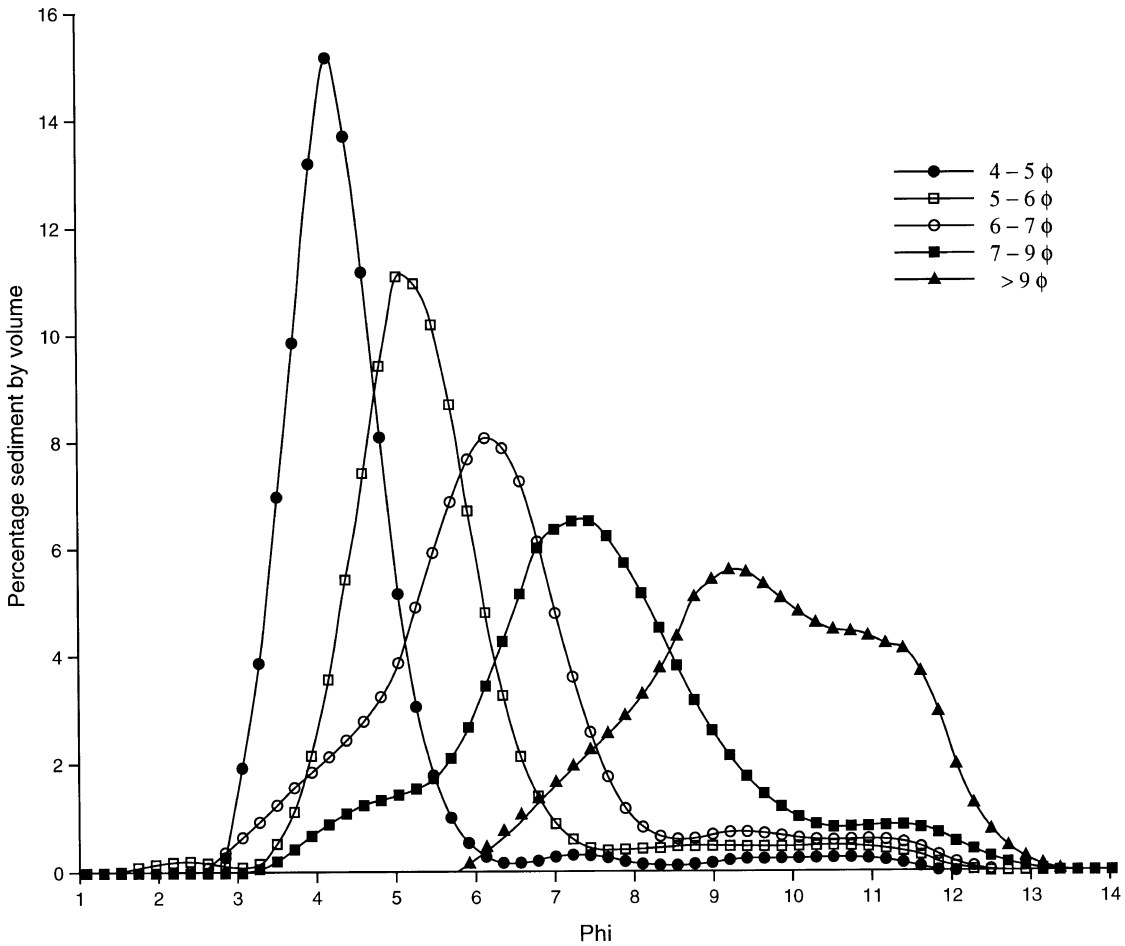

Figure 2. Grain size distribution of size fractions obtained from sample 1

Table III. Rank correlation coefficients between percentage of material by volume coarser than required size range and organic carbon content of size fractions from all samples. None are significant at 5 per cent confidence level

	Size fraction				
(phi)	4–5	5–6	6–7	7–9	>9
(μm)	32–63	16–32	8–16	2–8	<2
r_s coefficient	−0·64	−0·63	−0·19	−0·02	0·12

This lack of correlation is in agreement with evidence that organic matter is predominantly present in the form of coatings on fine-grained sediment in the marine environment rather than as individual particles (Mayer,1994).

The coarse tails illustrated in Figure 2 may therefore reflect certain inherent physical drawbacks associated with this method. It was noted that turbulence within the upper 10 cm of the settling tube persisted for at least 20 s after shaking the tube to ensure adequate sediment dispersal prior to fractionation, thereby delaying the onset of vertical settling and giving rise to the coarse tails observed in Figure 2. Furthermore, whilst the

accuracy of laser granulometry is not considered to be significantly affected by variations in particle shape (Matthews,1991), non-streamlined distortions of particle shape will serve to reduce particle settling velocity, which may further contribute to the presence of coarse tails in size fractions obtained using this technique.

Whilst the presence of coarse tails is undesirable, these will be of relatively little importance with regard to studies of contaminant concentrations in particle size fractions in comparison to the fine tails noted previously. Recent work has demonstrated strong correlations between the abundance of fine-grained sediment and various mineral magnetic properties in intertidal sediments (Clifton,1998; Oldfield and Yu,1994). Given the shortcomings of gravimetric procedures highlighted in the present study, it is recommended that normalization using sediment magnetic properties is undertaken in studies which utilize analysis of particle size fractions to infer the geochemical behaviour of trace metals or radionuclides.

ACKNOWLEDGEMENTS

The authors would like to acknowledge the support of BNFL for this project.

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